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# Accepted Manuscript

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# **Distribution and potential health impacts of microplastics and microrubbers in air and street dusts from Asaluyeh County, Iran**

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## Highlights

- 900 MPs and 250 MRs of varying size and shape were present in 15 g street dust
- MPs in air were fibrous but MRs were more difficult to characterise
- Acute exposure estimates via ingestion are 15 MP d<sup>-1</sup> and 7 MR d<sup>-1</sup> for children
- MPs and MRs exhibit oxidative potential that contributes to the toxicity of urban dust



## Abstract

While the distribution and effects of microplastics (MPs) have been extensively studied in aquatic systems, there exists little information on their occurrence in the terrestrial environment and their potential impacts on human health. In the present study, street dust and suspended dust were collected from the city and county of Asaluyeh, Iran. Samples were characterized by various microscopic techniques (fluorescence, polarized light, SEM) in order to quantify and classify MPs and microrubbers (MRs) in the urban and industrial environments that are potentially ingestible or inhalable by humans. In < 5-mm street dust retrieved from 15 sites, there were an average of 900 MPs and 250 MRs per 15 g of sample, with MPs exhibiting a range of

colours and sizes ( $< 100$  to  $> 1000 \mu\text{m}$ ). Most street dust samples were dominated by spherical and film-like particles and MRs largely made up of different sizes of black fragments and fibrous particulates. Airborne dust collected daily over an eight-day period at two locations revealed the ubiquity of fibrous MPs of sizes ranging from about  $2 \mu\text{m}$  to  $100 \mu\text{m}$  and an abundance of about  $1 \text{ per m}^{-3}$ . These samples contained small MR fragments whose precise characteristics were more difficult to define. Based on the median concentrations in street dust, estimates of acute exposure through ingestion are about  $5$  and  $15 \text{ MP d}^{-1}$  and  $2$  and  $7 \text{ MR d}^{-1}$  for construction workers and young children, respectively. Quantities of inhalable particulates were more difficult to define but the potential toxicity of MPs and MRs taken in by this route was evaluated from assays performed using particulates isolated from street dusts in the presence of an artificial lung fluid. Both types of particle exhibited oxidative potential, with MPs displaying consumptions of different antioxidants that were comparable with corresponding values for a reference urban particulate dust but lower than those for London ambient particulate matter. Thus, MPs and MRs contribute towards the health impacts of urban and industrial dusts but their precise roles remain unclear and warrant further study.

**Keywords:** Microplastics, Microrubbers, Dust, Exposure, Oxidative potential, Health risk

## 1. Introduction

Microplastics (MPs) are plastics whose primary dimension or diameter is less than  $5 \text{ mm}$  (Arthur et al. 2009). MPs may be either primary particulates, such as synthetic fibres from clothing and soft furnishings, microbeads in cosmetic and personal care products, plastic shot used in

60 industrial abrasives and resin pellets used by the plastic manufacturing industry, or secondary  
61 fragments that have been formed through the weathering or degradation of primary plastics  
62 (including primary MPs) (Alomar et al., 2016; Li et al., 2016). Because of their low density and  
63 persistence, coupled with inefficient or careless waste management and disposal practices, MPs  
64 are commonly observed in freshwater and marine environments and have been the subject of  
65 intense study over the past two decades (Ng and Obbard, 2006; Cole et al., 2014; Turner and  
66 Holmes, 2015). In aquatic systems, MPs can be mistaken for food and are inadvertently or  
67 deliberately ingested by a wide range of organisms, potentially causing blockages of and damage  
68 to the gastrointestinal tract and impacting on health and growth (Browne et al., 2015). Ingested  
69 MPs can also act as vectors for the bioaccumulation of chemical additives or monomers from the  
70 plastic matrix or contaminants adsorbed to the polymer surface (Batel et al., 2016). The direct or  
71 indirect (dietary) ingestion of MPs by commercially important species, coupled with possible  
72 translocation of micrometer-sized plastics has also recently raised concerns about the potential  
73 exposure of MPs to humans who regularly consume sea food (Rochman et al., 2015; Abbasi et  
74 al., 2018).

75 Despite their documented impacts on aquatic wildlife and potential dietary exposure to humans,  
76 very few studies have been undertaken on MPs in the terrestrial environment. In particular,  
77 information is lacking in the urban, industrial and domestic settings where the majority of  
78 primary environmental MPs are generated by a variety of processes, practices and industries  
79 (Dris et al., 2016). Here, MPs may accumulate in dusts and soils, but their low density ensures  
80 that they are readily suspended into the atmosphere. Thus, exposure may arise through the  
81 inadvertent ingestion of contaminated geosolids and the inhalation of fine, airborne materials.  
82 The impacts of MPs on human health are unknown but fibrous plastics of up to a few hundred

µm in length have been observed in malignant lung specimens (Pauly et al., 1998) and finer MPs inhaled or ingested are believed to be able to translocate to the circulatory system and to other organs (Wright and Kelly, 2017).

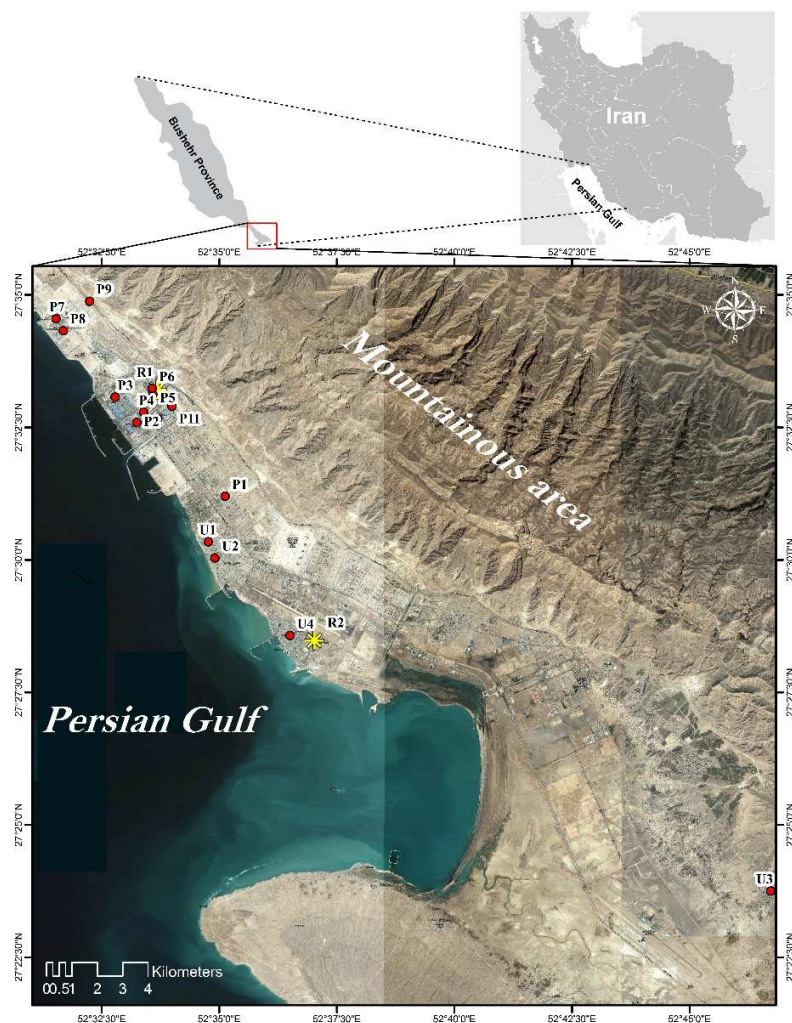
Recently, two studies have investigated the presence and distribution of MPs and microrubbers (MRs; a type of MP that is distinctly different in terms of its appearance, physical properties, deformation characteristics, sources and uses) in street dusts from urban environments in Iran. Specifically, Abbasi et al. (2017) demonstrated the widespread occurrence of MPs and MRs in dusts from Bushehr City using fluorescence microscopy, and Dehghani et al. (2017) characterized MPs isolated from dusts collected in Tehran by size, shape and colour using scanning electron microscopy (SEM). The latter study also estimated that outdoor exposures of MPs through incidental ingestion could be as high as several thousand per year for children and construction workers. Given the pervasiveness of MPs and MRs in street dusts, what is lacking is an evaluation of exposure arising from both deposited and airborne particulates, coupled with an assessment of the toxicity of particles that are potentially ingestible or inhalable. Accordingly, the present study combines the collection and microscopic characterisation of MPs and MRs in street dusts and suspended dusts from the city and county of Asaluyeh, southern Iran, with measures of their potential to form reactive oxygen species in artificial lung fluid. The findings are discussed in the context of the likely sources and environmental recycling of outdoor MPs and MRs and their contribution to the overall composition and toxicity of urban and industrial dusts.

## 2. Material and methods

### 2.1. Study area

Asaluyeh is a city and county located in the Bushehr province of southern Iran (Figure 1). Being the closest land point to the largest natural gas field in the world, the city and surroundings house the land-based facilities of the PSEEZ (Pars Special Energy/Economic Zone), a collection of different plants and refineries and associated heavy and light industries. The population of the city, including those working in the PSEEZ, is about 75,000. Bordered by the Persian Gulf to the southwest and mountains to the northeast, the dominant wind direction (southwesterly) ensures air is rather poorly ventilated and the city can experience air quality issues (Jafarigol et al., 2016).





**Figure 1** Location of the study area and sampling sites in Asaluyeh County. Red dots indicate street dust samples (P1 to P11 and U1 to U4) and yellow stars suspended dust samples (R1 and R2).

## 2.2. Sampling of MPs and MRs

A total of 31 samples (15 street dust samples and 16 suspended dust samples) were collected during the dry season (August 2017) from urban and industrial areas of the county at locations shown in Figure 1 and whose settings are described in Table S1. Approximately 100 g of street

dust was collected at 11 sites within the PSEEZ (P1 to P11) and at four urban sites (U1 to U4) from the road surface adjacent to the kerb. A metallic pan and wooden brush were used to carefully sweep material directly into air-tight, low-density polyethylene bags, with the pan and brush washed with filtered distilled water and dried between successive samples.

Two sampling stations were considered for (re-)suspended dusts, one in an industrial area within the PSEEZ (R1) and a second in an urban area (R2) (Figure 1 and Table S1). Samples of particulate matter (PM) were collected on eight successive days at both sites on polytetrafluoroethylene (PTFE) filter papers (46.2 mm in diameter and 2  $\mu\text{m}$  pore size; Tisch Scientific, USA) using an ECHO PM ambient filter sampler (TECORA, Italy). The dust collection system comprises a low-volume sampler unit and a filter changer with an intake tube and sampling head (inlet) to collect PM from air that is drawn through a size-selective inlet and through the filter media. Particulates with aerodynamic diameters less than the cut-point of the inlet are collected on the filter media, and here we focus on PM<sub>2.5</sub>, PM<sub>10</sub> and total suspended particulates (TSP). To avoid local disturbance from, for example, automobiles, the sampler was set up at a height of 3 to 4 m, and in accordance with US EPA reference methods for PM<sub>2.5</sub> and PM<sub>10</sub>, the sampling flow rate was set at 16.67 L min<sup>-1</sup> (EPA, 1997). After 24 h, filters were transferred to plastic petri dishes that had been washed with filtered water and returned to the laboratory for MP and MR counting.

### 2.3. Extraction and counting of MPs and MRs

For airborne MPs and MRs, the content on each filter was washed with deionized filtered water and carefully transferred to a glass beaker using a washed metallic needle before being dried in a sand bath at 80°C. Street dusts were air-dried for about 7 days before being passed through a 5-

mm stainless steel sieve to remove larger debris such as stones and vegetation. To remove organic matter from the street dust samples, 15 g aliquots of street dusts or the contents from the air filters were mixed with 35 mL of 30% H<sub>2</sub>O<sub>2</sub> for 8 days (and until bubble formation ceased). Residual H<sub>2</sub>O<sub>2</sub> on the dust particles was removed by vacuum-filtration through S&S filter papers (blue band, grade 589/3, 2 µm pore size) before filters were washed with distilled water and dried in a sand bath at 80°C. Fifty mL of a solution of NaI solution and of density 1.6 g cm<sup>-3</sup> was then added to each sample in a new beaker and the contents shaken for 5 min at 350 rpm before being allowed to settle for 90 min. The remaining supernatant was centrifuged for 3 min at 4000 rpm and then vacuum-filtered onto S&S blue band filter papers before being rinsed with distilled water to prevent the formation of NaI crystals. In order to capture all MPs and MRs, the process of density separation, centrifuging, and filtering was repeated three times through the same filter. Filters were dried at room temperature for a few days but in a metal cabinet in order to minimize contamination and subsequently transferred to petri dishes for counting.

For the identification (or confirmation) and isolation of MPs and MRs on the filters arising from the street dust samples and suspended ambient dust samples, binocular microscopy (Carl-Zeiss), polarized light microscopy (PLM) (Olympus BX41TF) and fluorescence microscopy (Olympus CX31) were used with commonly employed visual sorting methods (Stolte et al. 2015; Abbasi et al., 2017). Thus, physical characteristics, determined visually and microscopically and with the aid of tweezers, included form, hardness, gloss and colour (Hidalgo-Ruz et al. 2012), with MRs having a distinctively non-gloss black appearance, high elasticity and propensity to reversibly deform. MPs and MRs were classified according to colour as: white-transparent, yellow-orange, red-pink, blue-green or black-grey; according to shape as: fiber, film, fragment or spherule; and, with the aid of a 250 µm probe and ImageJ software, were categorized according to size in terms

of length or primary diameter as follows:  $L \leq 100 \mu\text{m}$ ;  $100 < L \leq 250 \mu\text{m}$ ;  $250 < L \leq 500 \mu\text{m}$ ;  $500 < L \leq 1000 \mu\text{m}$ ;  $1000 < L \leq 5000 \mu\text{m}$ .

Based on the optical microscopy results, the topography and elemental composition of selected MPs recovered from street dusts ( $n = 7$ ) was determined through high vacuum SEM/EDS. We used a Tescan VEGA 3 electron microscope (with a resolution of 2 nm at 20 kV) and an Oxford Instruments X-Max 50 silicon drift detector with AZtec and INCA software after samples that had been carefully brushed from the filters were mounted on double-sided adhesive carbon tabs on aluminium SEM stubs.

#### 2.4. QA/QC control

In order to prevent plastic and fiber contamination during the extraction phase of the dust samples in the laboratory, all reagents and distilled water were filtered through S&S blue band filters; working surfaces were thoroughly wiped with ethanol, all glassware was cleaned with distilled water and all windows and doors were closed. White cotton laboratory coats, single-use latex gloves and face masks were used throughout sample manipulation and processing and, when necessary, samples and containers were protected by Al foil. Two wide dishes full of filtered water were maintained in the laboratory for the duration of the extractions and subsequent analysis of filters arising from their filtration revealed no detectable MP or MR contamination under the working conditions employed. For replication purposes, five random street dust samples were recounted for MPs and MRs, yielding differences ranging from 5 to 17 and 2 to 8, respectively, from original counts.

#### 2.5. Oxidative potential (OP)

For the assays and analyses, all chemicals used were high grade (usually HPLC-grade), and were purchased from either the Sigma Chemical Company (UK) or VWR (UK). The OP assay was performed on MPs and MRs obtained from the street dust samples that had been separated during the initial counting phase. These were pooled and stored in parafilm-sealed petri dishes at 4 °C. Particles were resuspended in cryovials to 150 µg mL<sup>-1</sup> in 5% methanol in ultrapure chelex-100 resin-treated water (pH 7.0) with vigorous mixing and sonication (at an amplitude of 15 micron for 30 seconds). Resuspensions were then further diluted to 50 µg mL<sup>-1</sup> with chelex-treated water and stored at -70°C. To determine oxidative activity, 450 µL aliquots of the thawed suspensions were added into three 1.5 mL incubation tubes and incubated at 37°C for 10 min prior to the addition of 50 µL of synthetic respiratory tract lining fluid (RTLFL) (2mM each of ascorbic acid, AA, uric acid, UA, and reduced glutathione, GSH), resulting in a concentration of 200 µM per antioxidant. Triplicate samples were then incubated for 4 hours at 37 °C with constant mixing before incubation tubes were centrifuged at 13,000 rpm for 1 h at 4 °C. Aliquots of 16.7 µL of the RTLFL-exposed solutions were then added to 983.3 µL of cold (4 °C) 100 mM sodium phosphate buffer at pH 7.5 containing 1 mM EDTA in 1.5 mL Eppendorf centrifuge vials (for GSH analysis) and 50 µL aliquots of the solutions were added to 450 µL of cold 5.6% *meta*-phosphoric acid in 0.7 mL amber HPLC vials (for AA and UA analysis). All vials were immediately stored at -70°C and analysed within 1 week of storage. In addition to the MPs and MRs, particle-free controls, a negative control (M120; 50 µg mL<sup>-1</sup>) and a positive control (NIST 1648a urban particulate matter; 50 µg mL<sup>-1</sup>) were processed and stored likewise.

### 2.5.1. Determination of glutathione

This assay employs the technique of the GSSG reductase-DTNB linked assay based on the method of Baker et al. (1990). Thus, 50  $\mu$ L of the diluted, RTLf-exposed solutions were analysed in duplicate, together with glutathione standards, for total both total glutathione (GSX) and (following derivatization with 2-vinyl pyridine) for oxidised glutathione (GSSG) using a Spectramax190 (Molecular Devices, UK) with SoftMaxPro v4.8 software. The experimental minimum detection limit was 9  $\mu$ M GSSG, and reduced (GSH) glutathione was obtained by subtraction of GSSG value from GSX.

#### 2.5.2. Determination of AA and UA

This assay employs high performance liquid chromatography (HPLC) with an electrochemical detector based on the method of Iriyama et al. (1984) with modifications. Thus, aliquots of 20  $\mu$ L of acidified sample were injected onto a 150 x 4.6mm 5  $\mu$ m SphereClone ODS(2) column (Phenomenex, UK) and eluted with a 0.2 M  $K_2HPO_4$ - $H_3PO_4$  (pH 2.1) mobile phase containing 0.25 mM octanesulfonic acid. Eluates were analysed using a Gilson Scientific HPLC with Unipoint v5.1 software. Experimental minimum detection limits for ascorbic acid and uric acid were 7  $\mu$ M and 9  $\mu$ M, respectively, and final concentrations of AA and UA were calculated with external AA/UA standards which were run simultaneously.

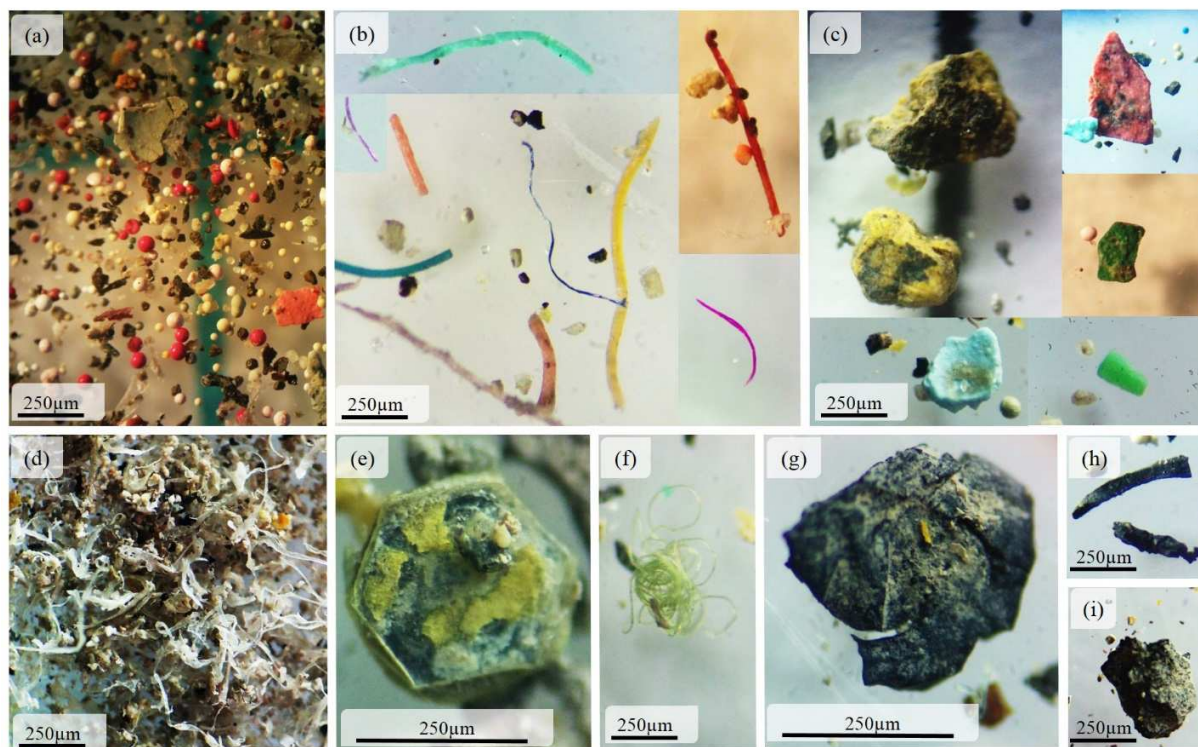
### 3. Results

#### 3.1. Characteristics of the MPs and MRs in street dust

Figure 2 exemplifies the types of microplastics (MPs) and microrubbers (MRs) that were observed as part of the present study and as captured by optical microscopy. With respect to the MPs, Figure 2a illustrates a wide variety of spherules whose diameters are less than 50  $\mu$ m,

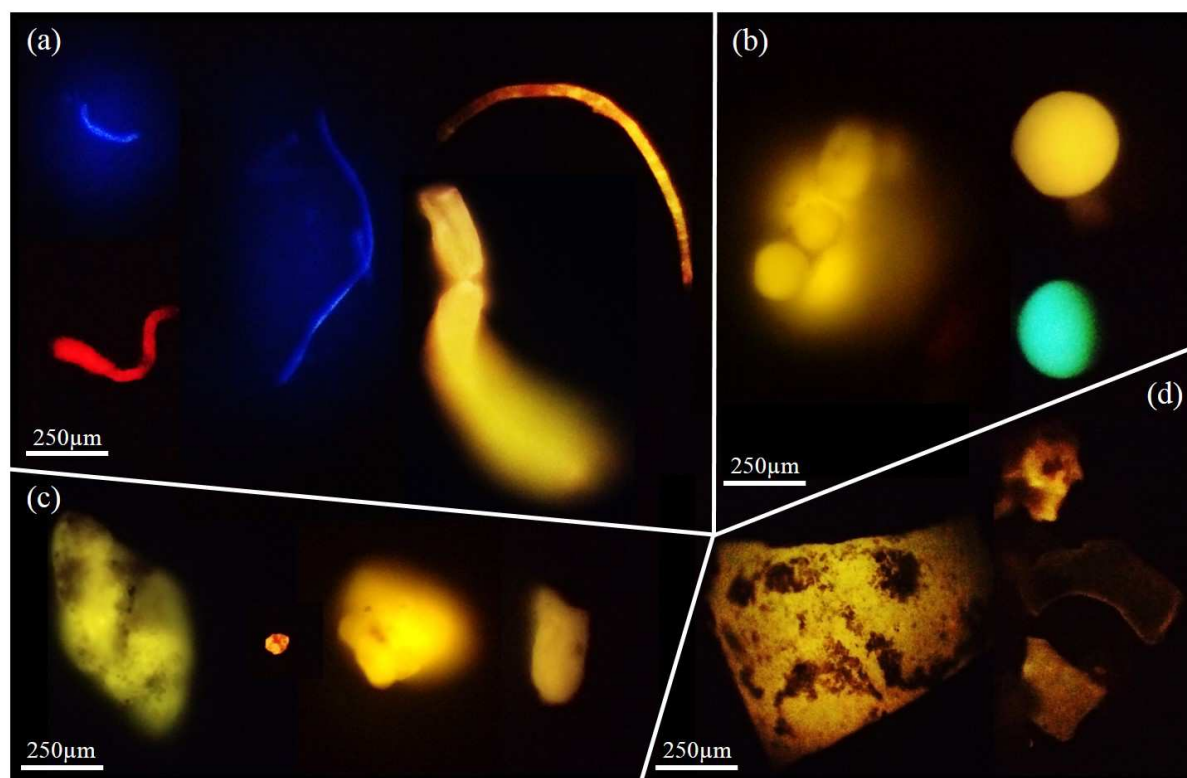
Figure 2b shows a number of fibres of different colour, length and diameter, Figure 2c illustrates a variety of colourful secondary fragments, Figures 2d and 2e show a number of irregular pieces and a single regular shape of film-like plastic, respectively, and Figure 2f displays a single, coiled fibrous thread. Regarding the MRs, Figures 2g, 2h and 2i illustrate a sheet-like layer, two fibres and an irregular fragment, respectively, all of which were black and of relatively low sheen. The presence of MPs (but not MRs) was also confirmed by fluorescence microscopy and polarized light microscopy. Regarding the former technique, MPs absorb ultraviolet light (300–400 nm) and radiate purple (400–450 nm) or blue (450–480 nm) fluorescence (Lei et al., 2006), as illustrated by the images of various fibres, spherules, fragments and films in Figure 3 (note that different coloured filters were used in the fluorescence microscopy in order to aid MP identification). With respect to the latter technique, sample rotation under polarized light results in characteristic colour changes (Abbasi et al., 2017), exemplified for various fibres, fragments and films in Figure 4.



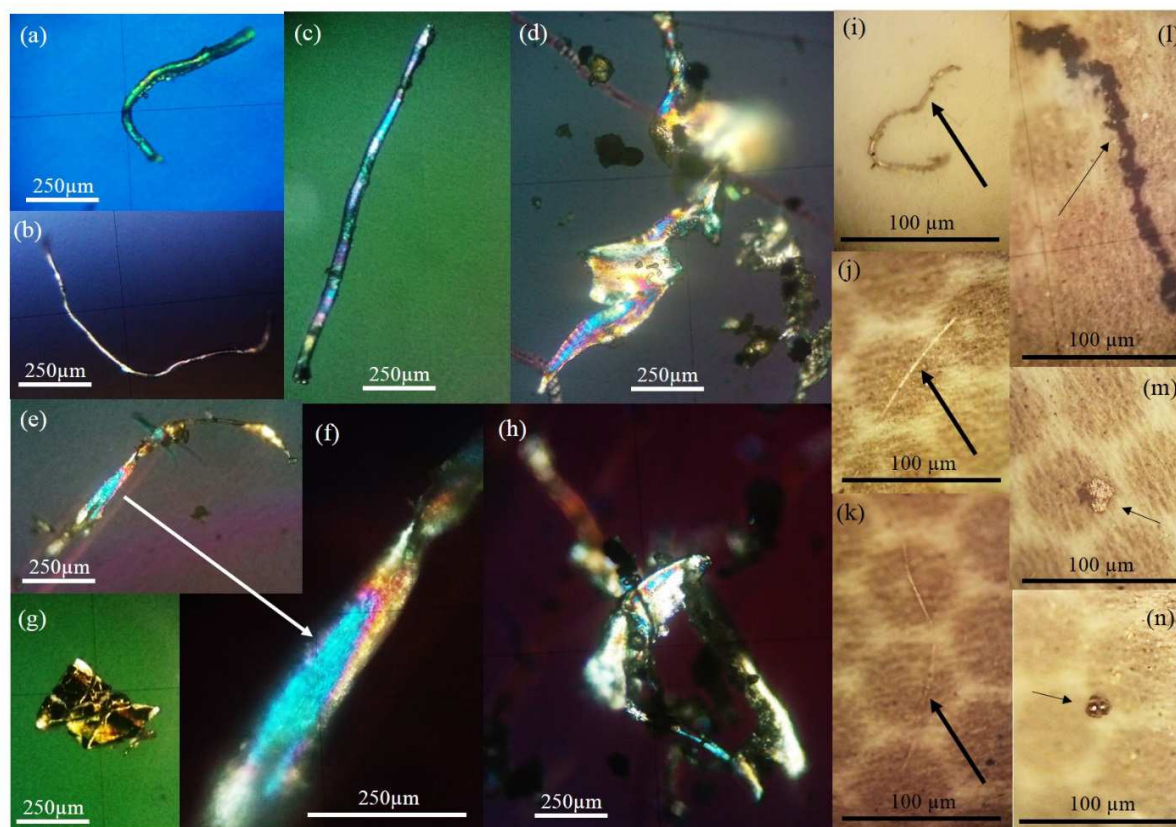


**Figure 2** Optical microscope images of different types of MP and MR. (a) Spherical MPs, (b) fibrous MPs, (c) fragmented MPs, (d) film-like MPs, (e) a single hexagonal film-like MP (probably from a primary source), (f) a thread-like MP, (g) film-like MRs, (h) fibrous MRs, and (i) a fragmented MR.



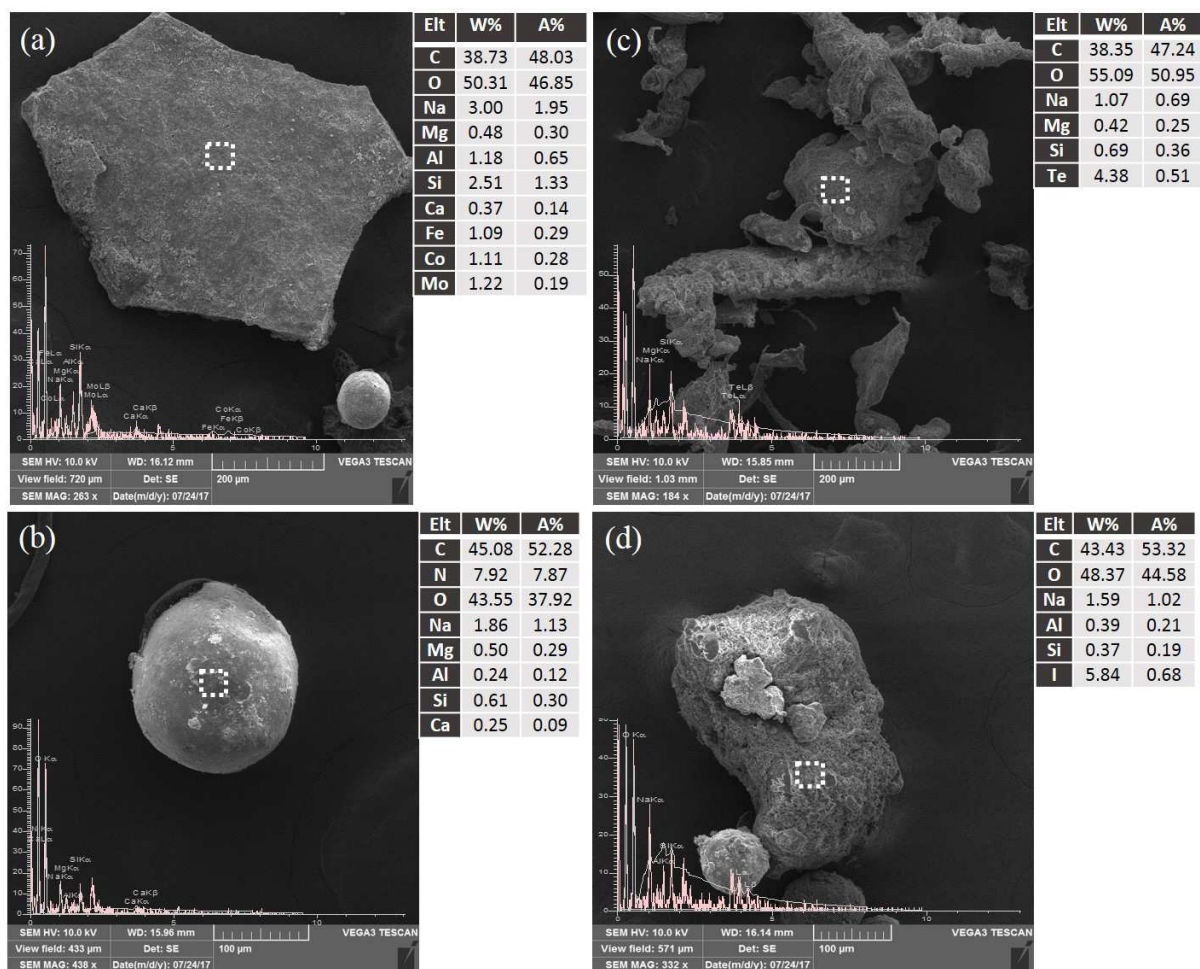


**Figure 3** Fluorescence microscopic images of (a) fibrous MPs, (b) spherical MPs, (c) fragmented MPs and (d) film-like MPs. Note that the colours reflect different the types of filtration employed during sample inspection.



**Figure 4** Polarized light microscopic images of (a, b and c) fibrous MPs, (d, e-f and h) film-like MPs (equivalent to Figures 3d and 5c), (g) a fragmented MP, (i, j and k) fibrous MPs on an air filter, (l) fibrous MRs on an air filter, and (m and n) metallic particles on an air filter.

Also, SEM/EDS was used to obtain high resolution images of the surface characteristics of selected MPs (21 images of 7 samples) and qualitative information about elemental composition, with results exemplified for a film, spherule and a number of fragments in Figure 5. Images illustrate a rather smooth surface with no fracturing, but evidence of mechanical and chemical weathering in the form of flaking and the presence of pits, grooves and irregular edges. Elemental results reveal that MPs are constructed mainly of C and O (and N for the spherule), with the presence of other elements likely reflecting contamination by extraneous solids such as dust and soil (Al, Ca, Si and Mg) or material used for sample preparation (Na and I).

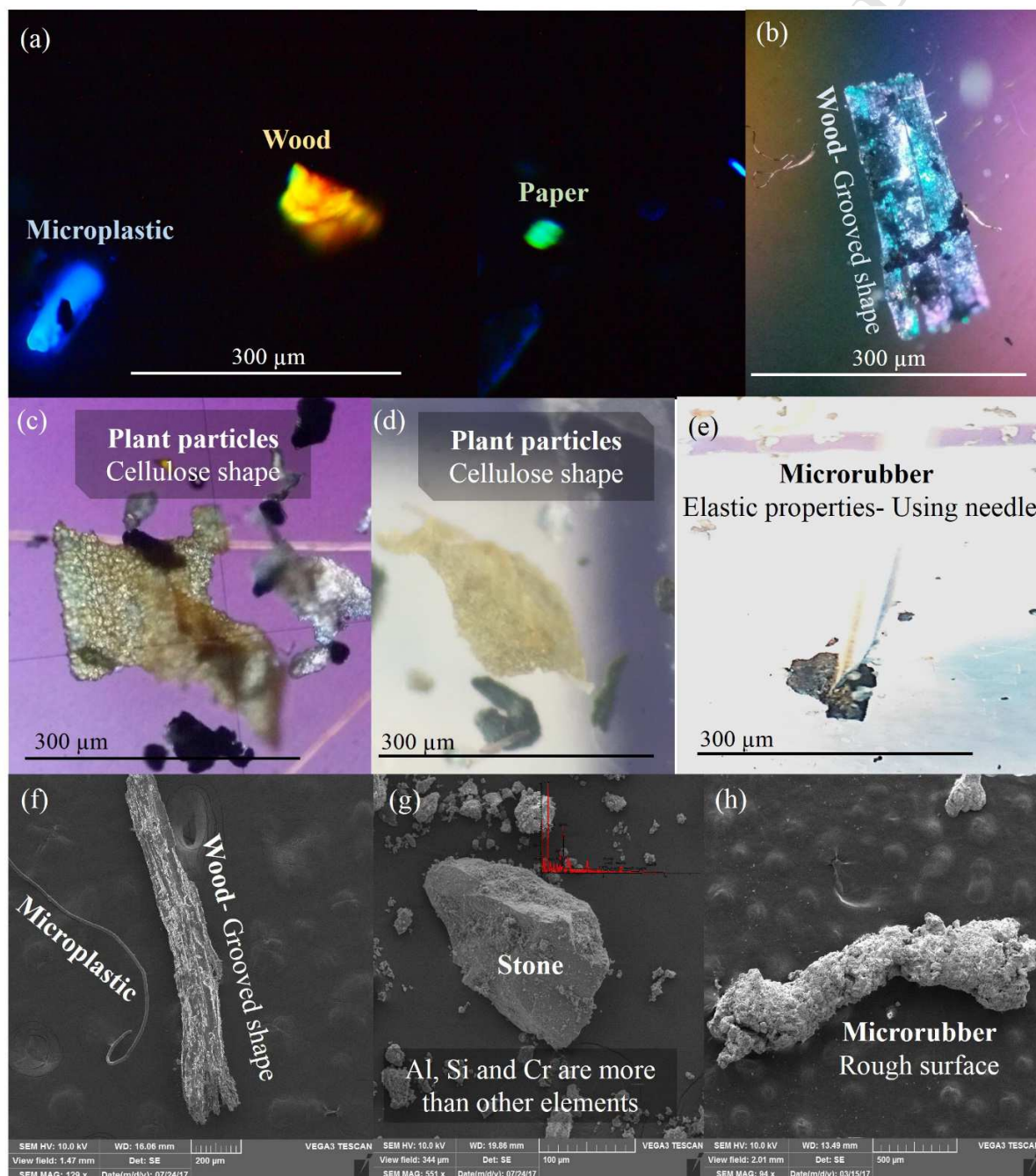


**Figure 5** SEM-EDS images of selected MPs. (a) film-like, (b) spherule (equivalent to Figure 2a), (c) film-like (equivalent to Figure 2d), (d) fragments (equivalent to Figure 2c).

Generally, among the particulate material remaining after treatment with  $\text{H}_2\text{O}_2$ , natural particles were identified (and eliminated from further consideration) by fluorescence and polarized light microscopy and by SEM/EDS according to specific characteristics and as exemplified in Figure 6. Thus, wood and paper particles showed as yellow or green under fluorescence microscopy when an ultra violet filter was used, while wood and plant material was characterized by distinctive shapes or grooving under polarized light microscopy; SEM/EDS revealed an



abundance of elements like Al, Si and Cr in inorganic particulates such as stones and, compared with MRs, a smoother surface (Figure 6).



**Figure 6** Examples of various different types of particulates captured by fluorescence microscopy (a), polarized light microscopy (b-e) and SEM/EDS (f, g and h).

### 3.2. Distribution and categorization of MPs and MRs in street dusts

Based on counting and visual characterisation by different microscopic techniques, Table S2 and Figure S1 show the number of MPs and MRs in the 15 street dust samples, along with their shape, colour and size classifications. In total, 13,132 MPs and 3691 MRs were identified among the samples considered. The highest concentrations of MPs were observed at sites P4 to P6 within the industrial PSEEZ region, with over 1000 particles per 15 g sample, and the lowest concentrations were observed at sites P8, P9 and U3 within both the industrial and urban areas and with about 50 particles per sample. The highest concentration of MRs was observed at P4, with over 1000 particles per 15 g, while the lowest concentrations were observed at sites P1, P8 and U3, with less than 40 particles per sample. Overall, the distributions of MPs and MRs were non-normal, with an average number of particles per sample of about 900 for MPs and 250 for MRs (or a ratio of about 3.5 MP: 1 MR) and an abundance of MPs exceeding that of MRs at all but two locations (P9 and U2).

The classification of particles by shape was very different between MPs and MRs. Thus, MPs were dominated by spherules (74%) and films (14%), while MRs were dominated by fragments (61%) and fibres (36%) and with only one sphere-like particle identified overall. In general, there appeared to be more film-like, spherical and fragmented MPs in the industrial region and more fibrous MPs in the urban areas. Despite these observations, however, a non-parametric Kruskal-Wallis test revealed no significant difference ( $p = 0.793$ ) between different shapes of MPs among individual sampling stations.

Regarding the colour classification of MPs (MRs are defined as black), white-transparent was most abundant overall (about 66%), with a roughly even split among the remaining colours. This percentage distribution was similar in the fibre and spherule categories, but among film MPs white-transparent particles comprised over 90% of the total population and in the fragment category the relative abundance of white-transparent was only about 45%, with red-pink and blue-green MPs representing about 20% each. The principal difference between the urban and industrial regions appeared to be a distinctively higher proportion of black MPs among all shape categories, and in particular in spherules, in the latter setting.

With respect to the size of MPs, the distribution overall was positively skewed, with more than 75% of particles occurring in the  $< 100 \mu\text{m}$  category and progressively smaller percentages encountered with increasing size. This distribution was accentuated among the spherules, with around 98% of particles occurring in the smallest size category and only one MP encountered between 500 and 1000  $\mu\text{m}$  and one above 1000  $\mu\text{m}$ . Among the remaining shape types, the size distribution was more normally distributed, with the highest percentage of MPs encountered within the 100 to 250  $\mu\text{m}$  size range. This type of distribution was also a characteristic of all shape types of MR with the exception of spherules, where only a single particle was observed overall and that occurred in the smallest size classification.

### *3.3. MPs and MRs in suspended dusts*

The number, colour and size classification of suspended MPs collected on the air filters during the eight-day sampling periods at R1 and R2 are shown in Table 1. Note that the data are limited to fibrous materials since only six non-fibrous plastics (three fragments and three film-like plastics) were retrieved over the whole sampling campaign. In addition to MPs, other particles

observed on the sample filters included MRs of different sizes but mainly fragmented and occasionally fibrous and particles exhibiting a metallic appearance (illustrated under binocular microscopy in Figure 4m and 4n).

**Table 1:** Number, size and colour classification of fibrous MPs collected on air filters over 8-day periods at sites R1 and R2.

sample location/date	sampler inlet	air volume (m <sup>3</sup> )	MP size (µm)	white/transp.	yellow/orange	red/pink	blue/green	black/grey
<i>industrial, R1</i>								
06/08/2017	TSP	23.60	L < 2.5	3		2		1
			2.5 < L < 10	5				1
			L > 10	5		1		
07/08/2017	PM10	23.91	L < 2.5	2		1		
			2.5 < L < 10	5				
			L > 10	6	1			
08/08/2017	PM2.5	23.81	L < 2.5					
			2.5 < L < 10					
			L > 10					
22/08/2017	PM10	23.77	L < 2.5	1				
			2.5 < L < 10	2				
			L > 10	5				1
23/08/2017	PM10	23.77	L < 2.5	1				2
			2.5 < L < 10	1		2		
			L > 10			4		1
24/08/2017	PM10	23.92	L < 2.5	1	1	2		
			2.5 < L < 10	3		3		1
			L > 10	3				
27/08/2017	PM10	23.89	L < 2.5	1				
			2.5 < L < 10	5		1		
			L > 10	2			1	
31/07/2017	PM10	23.67	L < 2.5	2				1
			2.5 < L < 10	4		1		
			L > 10	15				1
<i>total industrial</i>				72	2	17	1	9
<i>urban, R2</i>								
06/08/2017	TSP	23.67	L < 2.5	2				
			2.5 < L < 10	4				
			L > 10	3	1	3		2
07/08/2017	PM10	23.88	L < 2.5	3		2		
			2.5 < L < 10	1				
			L > 10	5		1		3
08/08/2017	PM2.5	23.93	L < 2.5					
			2.5 < L < 10	1				
			L > 10					
22/08/2017	PM10	23.71	L < 2.5	4		2		3
			2.5 < L < 10	6				
			L > 10	4		3		
23/08/2017	PM10	23.79	L < 2.5	5		4		7
			2.5 < L < 10			3		1
			L > 10	3				2
24/08/2017	PM10	23.92	L < 2.5			6	1	4
			2.5 < L < 10	1		3	1	4
			L > 10	4				1
27/08/2017	PM10	23.93	L < 2.5	1		1		1
			2.5 < L < 10	1				2
			L > 10	1		1		
31/07/2017	PM10	23.92	L < 2.5	1				2
			2.5 < L < 10					1
			L > 10	3		1		
<i>total urban</i>				53	1	30	2	33



The aerodynamic diameter of the sampler inlet had an impact on the number of MPs retrieved, with relatively few particles observed on samples taken with the smallest (PM<sub>2.5</sub>) diameter. However, it is important to mention that the number of MPs less than 2.5  $\mu\text{m}$  may have been underestimated because of difficulties in visually detecting particles of this size (2  $\mu\text{m}$  has been considered as a minimum detectable size). Moreover, the size distributions observed may not accurately reflect inlet diameter because fibres (the dominant shape of suspended MPs) are typically characterized by both a relatively long and relatively short dimension and their collection or trapping according to size is partly dependent on their orientation with respect to the inlet port. Note that after initial trials with different inlet sizes, the samplers were fixed at PM<sub>10</sub> throughout the remaining, 5-day sampling periods.

The total number of fibrous, airborne MPs collected during the sampling campaign was 220, with a roughly equal split between the urban and industrial regions. When normalized to airflow, and excluding samples collected with the PM<sub>2.5</sub> intake, the number of MPs per  $\text{m}^3$  ranged from about 0.3 to 1.1. The dominant colour of these MPs was white-transparent, comprising in excess of 70% and 53% of the total MPs in the industrial and urban settings, respectively. Only three blue-green and yellow-orange MPs were observed throughout, and the principal colour difference between the two regions was black, with 33 counted in the industrial zone and 9 in the urban environment.

MRs were clearly visible on all air filters inspected microscopically. However, particles were always fine and sometimes brittle, imposing constraints on accurate quantification and classification. For these reasons, airborne MRs are subsequently referred to but their precise details are not addressed.

#### 3.4. Differences between street and suspended dusts

The main difference in MPs between the street and suspended dusts is the variety of shapes. Thus, street dusts are characterized by a heterogeneous assortment of primary and secondary plastics of different size while airborne dusts are limited to fine, fibrous plastics. With respect to street dusts, fibrous material constitutes only about 6% of MPs in the industrial setting but more than 30% of MPs in the urban environment. Despite these differences, the colour distribution of fibrous MPs is rather similar between street and suspended dusts, being dominated by white-transparent material and very few yellow-orange or blue-green fibres. This observation suggests that fibrous material in street and suspended dusts has the same source that is likely related to clothing and soft furnishings.

The presence of microfibers in the atmosphere may be attributed to the low mass-high surface area and more ready suspension of fibre-like plastics compared with spheres, fragments and films, with variations over the sampling period therefore reflecting temporal variations in local meteorological and environmental conditions. The number of MPs suspended was not clearly related to any single meteorological measure (e.g. wind speed, wind direction, humidity, temperature) but was correlated with the concentration of dust suspended in the local atmosphere (in  $\text{g m}^{-3}$ ) and determined from the mass of material retained on filters ( $r = 0.529$ ,  $p = 0.0349$ ;  $n = 16$ ). This association suggests common mechanisms of resuspension and transportation of airborne dust and MPs (through, for instance, local meteorology, thermal heating and wind modulation).

#### 3.5. Human intake of MPs and MRs and their potential health risks

The number of ingested MPs and MRs per day was calculated under various scenarios based on average dust intake values of 100 and 200 mg day<sup>-1</sup> for adults and children, respectively (US EPA, 2002) and mean particle ingestion from acute exposure in children of 1 g soil per outdoor day and in construction or outdoor workers of 330 mg per day (Harris and Harper, 2004; Dehghani et al., 2017). Computed daily ingestion of MPs and MRs arising from street dusts at each site, shown in Table 2, reveal an intake of MPs under normal conditions range from about 0.3 to 50 day<sup>-1</sup> (median = 1.5 day<sup>-1</sup>) and from about 0.7 to 100 day<sup>-1</sup> (median = 3.0 day<sup>-1</sup>) for adults and children, respectively, with corresponding approximate ranges of MRs of 0.2 to 9 day<sup>-1</sup> (median = 0.7 day<sup>-1</sup>) and 0.4 to 18 day<sup>-1</sup> (median = 1.4 day<sup>-1</sup>). Under acute exposure, the estimated intake of MPs and MRs are 5 or 3.3 times greater for adults and children, respectively.

**Table 2:** Estimated number of daily ingested MPs and MRs at each sampling location by adults and children under two exposure scenarios (NE = normal exposure; AE = acute exposure).

sampling site	MPs 15 g <sup>-1</sup> street dust	MPs adults		MPs children		MRs 15 g <sup>-1</sup> street dust	MRs adults		MRs children	
		NE, day <sup>-1</sup>	AE, day <sup>-1</sup>	NE, day <sup>-1</sup>	AE, day <sup>-1</sup>		NE, day <sup>-1</sup>	AE, day <sup>-1</sup>	NE, day <sup>-1</sup>	AE, day <sup>-1</sup>
P1	115	0.8	2.5	1.5	7.7	39	0.3	0.9	0.5	2.6
P2	82	0.5	1.8	1.1	5.5	47	0.3	1.0	0.6	3.1
P3	301	2.0	6.6	4.0	20.1	106	0.7	2.3	1.4	7.1
P4	1295	8.6	28.5	17.3	86.3	1325	8.8	29.2	17.7	88.3
P5	1428	9.5	31.4	19	95.2	393	2.6	8.6	5.2	26.2
P6	7748	51.7	170.5	103.3	516.5	162	1.1	3.6	2.2	10.8
P7	398	2.7	8.8	5.3	26.5	223	1.5	4.9	3	14.9
P8	59	0.4	1.3	0.8	3.9	37	0.2	0.8	0.5	2.5
P9	52	0.3	1.1	0.7	3.5	104	0.7	2.3	1.4	6.9
P10	486	3.2	10.7	6.5	32.4	130	0.9	2.9	1.7	8.7
P11	627	4.2	13.8	8.4	41.8	52	0.3	1.1	0.7	3.5
U1	109	0.7	2.4	1.5	7.3	107	0.7	2.4	1.4	7.1
U2	158	1.1	3.5	2.1	10.5	891	5.9	19.6	11.9	59.4
U3	52	0.3	1.1	0.7	3.5	28	0.2	0.6	0.4	1.9
U4	222	1.5	4.9	3.0	14.8	47	0.3	1	0.6	3.1
median	222	1.5	4.9	3.0	14.8	106	0.7	2.3	1.4	7.1

The results of the antioxidant depletion measurements (oxidative potential; OP) of MPs and MRs isolated from the street dust samples are summarised in Table 3. Here, the percentages of

antioxidants (AA, UA and GSH) consumed after 4 h incubation are shown after correction for a particle-free control, along with percentages of OP due to AA and GSH expressed per  $\mu\text{g}$  of material and the total OP per  $\mu\text{g}$  arising from AA and GSH. (No observable reaction of MRs with UA and only limited reaction of MPs with UA were observed.) By comparison, the negative control (M120) displayed no reactivity with the antioxidants and the positive control (NIST 1648a) displayed 30% consumption of AA and 18% consumption of GSH. It is important to appreciate that these measures relate to both the inherent properties of the MPs and MRs themselves (including any dyes and additives) as well as any organic and inorganic contaminants adsorbed to their surfaces. Moreover, since the RTLF model mimics the interaction with antioxidants naturally found in the surface of the lung, an implicit assumption of the results is that particles tested are inhalable. Clearly, MPs and MRs in street dusts span a broader range of size and shape than airborne dusts (Table S1 and Table 1) but it is presumed that the two types of dust exhibit similar oxidative characteristics through common sources and physico-chemical characteristics. Within these limitations and assumptions, the results indicate that, in all cases and according to both measures, MPs, on average, consume greater quantities of antioxidants than MRs.

**Table 3:** Mean ( $\pm$  one standard deviation;  $n = 3$  for each sample) antioxidant consumption, as a percentage and normalized to mass, for a 4 h incubation of MPs and MRs.

	MPs	MRs
AA, %	17.0 $\pm$ 0.9	2.9 $\pm$ 1.2
UA, %	0.2 $\pm$ 1.5	nd
GSH, %	12.2 $\pm$ 0.8	8.6 $\pm$ 2.6
OP <sub>AA</sub> / $\mu\text{g}$	0.34 $\pm$ 0.02	0.06 $\pm$ 0.02
OP <sub>GSH</sub> / $\mu\text{g}$	0.24 $\pm$ 0.02	0.17 $\pm$ 0.05
OP <sub>total</sub> / $\mu\text{g}$	0.58	0.19

#### 4. Discussion

There has been a great deal of attention paid to the impacts of well-defined or controlled MPs on aquatic organisms that ingest material. For example, ingested MPs have been shown to adversely affect growth, accumulate in the liver resulting in inflammation, and act as a vector for the accumulation of adsorbed chemical contaminants (Avio et al., 2015; Lu et al., 2016; Zhang et al., 2017). Very few studies, however, have been undertaken on the occurrence of MPs (and MRs) in the urban setting and the implications for human exposure through either ingestion or inhalation (Abbasi et al., 2017; Dehghani et al., 2017). The results of the present study reveal that street dusts from urban and industrial regions of coastal Iran contain a heterogeneous array of particles of different shape, size and colour, with an abundance of between about 3 and a few hundred MPs and about 2 and 100 MRs per g. MPs are likely derived from a multitude of sources, including industrial emissions, synthetic textiles, decomposing litter, waste disposal and agricultural practices (Prata, 2018), while MRs are likely derived largely from vehicle tire-wear (and, possibly, footwear) with a distribution that reflects traffic density and driving habits (Turner and Hallett, 2012). In the atmosphere, (re-)suspended particles appear to be limited to relatively few MRs of various shapes, and MPs that are largely of a fibrous nature whose abundance is around 1 per  $\text{m}^{-3}$  or less. Previous studies have confirmed the dominance of synthetic fibre-like particles in the atmospheric microplastic pool of Paris (Dris et al., 2015; 2016) and Dongguan City (China) (Cai et al., 2017) and have suggested sources that include clothing, household soft furnishings, vehicle interiors and certain industrial processes and practices.

Thus far, human ingestion of MPs has focused on dietary exposure, where MPs may be found in a variety of foodstuffs that include salt (Yang et al., 2015), beverages (Liebezeit and Liebezeit, 2014) and seafood (Li et al., 2015; Abbasi et al., 2018). The current study, however, demonstrates the potential a wider variety of MPs and MRs to be ingested inadvertently and through the incidental exposure of dusts, a vector that is likely most significant for certain occupations and for young children. Regarding the latter, exposure estimates based on the present results range from about 3 to > 500 MPs per day and 2 to 90 MRs per day. By comparison, and based on recent observations in fish and shellfish tissue, the average daily ingestion of MPs through the consumption of seafood in Iran is around 5 (Abbasi et al., 2018). Exposure to airborne suspended MPs and MRs occurs through inhalation. Studies of workers in factories processing synthetic fibrous materials (e.g. nylon, polyamide) or plastics have demonstrated increase incidences of various respiratory diseases, albeit at MP concentrations far higher than levels observed outdoors (Prata, 2018). Significantly, however, microfibers on the order of a few hundred  $\mu\text{m}$  have been observed in the deep lung of malignant tissue suggesting that some fibres are able to avoid clearance and that their persistence has the potential to induce acute and chronic inflammation (Pauly et al., 1998). The MPs and MRs retrieved from street dusts in the present study are assumed to be representative of the stock of suspended (inhalable) particles in terms of their oxidative potential, albeit of a different size and shape. Results indicate a greater OP for MPs than MRs with percentage consumptions of AA and GSH of 17 and 12.2 that are comparable with corresponding values for a reference urban dust (NIST 1648a; 30 and 18, respectively). Result for  $\text{OP}_{\text{AA}}$  on a mass basis, however ( $= 0.34 \mu\text{g}^{-1}$ ), are an order of magnitude lower than equivalent values for roadside ambient PM samples from the city of London ( $5.32 \pm 4.52 \mu\text{g}^{-1}$ ;

Godri et al., 2011). Nevertheless, it is clear from this study that MPs and MRs make a contribution to the overall toxicity and morbidity of urban and industrial dusts whose significance is likely to be proportional to their relative abundance in suspended dust and that is predicted to increase as the use of plastics rises or as the current stock of material weathers and degrades. This contribution may be more important to vulnerable individuals such as very young children, the elderly and those suffering from respiratory problems.

Small, submicrometer MPs interacting with the respiratory (and gastrointestinal) epithelium also have the potential to be translocated to the circulatory system and to other organs through diffusion or cellular penetration (Prata, 2018). While particles of this dimension were not detected in our samples, the breakdown, weathering and abrasion of materials in the outdoor environment suggests that MPs and MRs of these dimensions are feasible. Moreover, larger particles (up to 130  $\mu\text{m}$ ) also have the potential to be taken up mechanically in the gastrointestinal environment through gaps in the epithelium at the villus tips through paracellular persorption (Wright and Kelly, 2017). Although plastic particles from prosthetic implants have been identified in the human circulatory system and different secondary tissues (Urban et al., 2000), their precise reactions and impacts are unknown. However, the immunological response appears to exhibit some dependence on the chemical composition of the material (for instance, polyethylene terephthalate is more harmful than polyethylene) and is likely to be related to various physical factors, like size and shape, and chemical factors, like the presence and migratability of monomers and endogenous additives (Wright and Kelly, 2017).

The presence of MPs and MRs in street and suspended dusts of urban and industrial settings also raises the possibility of contaminating environments and ecosystems farther afield. Presumably, fine, airborne MPs of mainly a fibrous nature have the potential for long-range transport and the

direct contamination (or re-contamination) of agricultural soil and surface water bodies when washed out. Larger and more diverse particles in street dust whose densities are lower than those of mineral grains have the potential to impact these systems indirectly via contamination of and transportation with street runoff and sewage sludge-based fertilisers.

The observations herein also have implications for exposure through ingestion and inhalation in the indoor environment. Thus, exogenous, industrial and urban MPs and MRs may be readily tracked into the household through shoes or enter ventilated buildings as airborne particulates. Conversely, the indoor environment itself has been implicated as a significant source of external, airborne, fibrous MPs that are derived from clothing and soft furnishings of households and vehicles interiors. In a recent comparison of interior and exterior suspended particulates, Dris et al. (2017) showed that fibrous material was more abundant indoors (up to 60 per m<sup>-3</sup>) but was characterized by smaller (shorter) particles overall. The majority of indoor fibres consisted of natural products, like cotton or other cellulose-based materials, but significant quantities consisted of petroleum-based products, and in particular polypropylene.

## 5. Conclusions

Deposited and airborne MPs and MRs are potentially significant contaminants in the urban and industrial settings that have received little attention. The present study has shown that street dusts in an industrialized and urbanized district of coastal Iran contain an average of about 900 MPs and 250 MRs per 15 g sample. MPs are a heterogeneous array of fibres, fragments, spherules and films of various colour and size that are derived from a multitude of domestic, vehicular and industrial sources. In contrast MRs are less variable in terms of shape and colour and appear to be derived largely from tire wear. In the local atmosphere, MPs are dominated by fibres at



concentrations of up to about  $1 \text{ m}^{-3}$  and that are mainly coloured white-transparent and, while MRs were observed, they were difficult to quantify and characterize. Both MPs and MRs exhibit oxidative potential, with the former displaying percentage consumptions of different antioxidants that are comparable with corresponding values for a reference urban particulate dust. The health impacts of MPs and MRs in the urban and industrial settings arising from ingestion or inhalation are unclear but warrant further investigation.

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**Figure S1.** Size classification of MPs and MRs in the street dust samples

**Table S1.** Land use characteristics of the sampling stations in the industrial and urban regions of Asaluyeh County.

**Table S2.** Number and characteristics of MPs and MRs from street dusts of Asaluyeh County.

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**Table 1:** Number, size and colour classification of fibrous MPs collected on air filters over 8-day periods at sites R1 and R2.

sample location/date	sampler inlet	air volume (m <sup>3</sup> )	MP size (μm)	white/transp.	yellow/orange	red/pink	blue/green	black/grey
<i>industrial, R1</i>								
8/6/2017	TSP	23.60	L < 25	3		2		1
			25 < L < 100	5				1
			L > 100	5		1		
8/7/2017	PM10	23.91	L < 25	2		1		
			25 < L < 100	5				
			L > 100	6	1			
8/8/2017	PM2.5	23.81	L < 25					
			25 < L < 100					
			L > 100					
8/22/2017	PM10	23.77	L < 25	1				
			25 < L < 100	2				
			L > 100	5				1
8/23/2017	PM10	23.77	L < 25	1				2
			25 < L < 100	1		2		
			L > 100			4		1
8/24/2017	PM10	23.92	L < 25	1	1	2		
			25 < L < 100	3		3		1
			L > 100	3				
8/27/2017	PM10	23.89	L < 25	1				
			25 < L < 100	5		1		



			L > 100	2			1	
7/31/2017	PM10	23.67	L < 25	2				1
			25 < L < 100	4		1		
			L > 100	15				1
<i>total industrial</i>				72	2	17	1	9
<i>urban, R2</i>								
8/6/2017	TSP	23.67	L < 25	2				
			25 < L < 100	4				
			L > 100	3	1	3		2
8/7/2017	PM10	23.88	L < 25	3		2		
			25 < L < 100	1				
			L > 100	5		1		3
8/8/2017	PM2.5	23.93	L < 25					
			25 < L < 100	1				
			L > 100					
8/22/2017	PM10	23.71	L < 25	4		2		3
			25 < L < 100	6				
			L > 100	4		3		
8/23/2017	PM10	23.79	L < 25	5		4		7
			25 < L < 100			3		1
			L > 100	3				2
8/24/2017	PM10	23.92	L < 25			6	1	4
			25 < L < 100	1		3	1	4

			L > 100	4				1
8/27/2017	PM10	23.93	L < 25	1		1		1
			25 < L < 100	1			2	
			L > 100	1		1		
7/31/2017	PM10	23.92	L < 25	1				2
			25 < L < 100				1	
			L > 100	3		1		
total urban				53	1	30	2	33

**Table 2:** Estimated number of daily ingested MPs and MRs at each sampling location by adults and children under two exposure scenarios (NE = normal exposure; AE = acute exposure).

Sampling site	MPs 15 g <sup>-1</sup> street dust	MPs adults		MPs children		MRs 15 g <sup>-1</sup> street dust	MRs adults		MRs children	
		NE, day <sup>-1</sup>	AE, day <sup>-1</sup>	NE, day <sup>-1</sup>	AE, day <sup>-1</sup>		NE, day <sup>-1</sup>	AE, day <sup>-1</sup>	NE, day <sup>-1</sup>	AE, day <sup>-1</sup>
P1	115	0.8	2.5	1.5	7.7	39	0.3	0.9	0.5	2.6
P2	82	0.5	1.8	1.1	5.5	47	0.3	1.0	0.6	3.1
P3	301	2.0	6.6	4.0	20.1	106	0.7	2.3	1.4	7.1
P4	1295	8.6	28.5	17.3	86.3	1325	8.8	29.2	17.7	88.3
P5	1428	9.5	31.4	19	95.2	393	2.6	8.6	5.2	26.2
P6	7748	51.7	170.5	103.3	516.5	162	1.1	3.6	2.2	10.8
P7	398	2.7	8.8	5.3	26.5	223	1.5	4.9	3	14.9
P8	59	0.4	1.3	0.8	3.9	37	0.2	0.8	0.5	2.5
P9	52	0.3	1.1	0.7	3.5	104	0.7	2.3	1.4	6.9
P10	486	3.2	10.7	6.5	32.4	130	0.9	2.9	1.7	8.7
P11	627	4.2	13.8	8.4	41.8	52	0.3	1.1	0.7	3.5
U1	109	0.7	2.4	1.5	7.3	107	0.7	2.4	1.4	7.1
U2	158	1.1	3.5	2.1	10.5	891	5.9	19.6	11.9	59.4
U3	52	0.3	1.1	0.7	3.5	28	0.2	0.6	0.4	1.9
U4	222	1.5	4.9	3.0	14.8	47	0.3	1	0.6	3.1
median	222	1.5	4.9	3.0	14.8	106	0.7	2.3	1.4	7.1

**Table 3:** Mean ( $\pm$  one standard deviation;  $n = 3$  for each sample) antioxidant consumption, as a percentage and normalized to mass, for a 4 h incubation of MPs and MRs.

	MPs	MRs
AA, %	17.0 $\pm$ 0.9	2.9 $\pm$ 1.2
UA, %	0.2 $\pm$ 1.5	nd
GSH, %	12.2 $\pm$ 0.8	8.6 $\pm$ 2.6
OP <sub>AA</sub> / $\mu$ g	0.34 $\pm$ 0.02	0.06 $\pm$ 0.02
OP <sub>GSH</sub> / $\mu$ g	0.24 $\pm$ 0.02	0.17 $\pm$ 0.05
OP <sub>total</sub> / $\mu$ g	0.58	0.19